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# Effect of Chemical Treatments and Coupling Agents on the Properties of Unidirectional Jute Fiber Reinforced Polypropylene Composite

(Kesan Rawatan Kimia dan Agen Gandingan ke Atas Ciri-ciri Gentian Jut Searah Diperkukuh Komposit Polipropelina)

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ABSTRACT

*Due to different chemical nature of jute and polypropylene, their blended composite materials often show failures behavior when applied in load bearing purposes. The aim of the research is to improve the properties of unidirectional jute fiber (JF) reinforced polypropylene (PP) composite. Unidirectional fiber composite was produced by simple hot press molding method according to weight percentage of 30wt% filler (JF), 2wt% coupling agent (compatibilizer) degraded PP (DgPP) and maleated PP (MAPP) and 68 wt% matrix (PP). To improve the interfacial interactions between the fiber and PP, the fiber was chemically modified with 10w/v% NaOH, 6w/v% dicumylperoxide (DCPO), 0.05 w/v% $K_2Cr_2O_7$ and 5 w/v% $H_2O_2$ solution. The water absorption, tensile and thermal properties of raw and surface treated fibers composites were compared in particular coupling agent. It has found that the fiber treatments and addition of coupling agents, composites showed better properties. Among the composites, 10 wt/v% NaOH treated jute reinforced PP composite by using MAPP compatibilizer was presented better water absorption and mechanical properties. The positive change of the treated fiber composites was also found in case of the thermal stability that investigated by the TG, DTA and DTG tests.*

*Keywords: Jute fiber (filler); Polypropylene (matrix); Composites; Coupling agents; Properties*

## INTRODUCTION

The development of composites furnished by ligno-cellulosic fibers as reinforcing materials of thermoplastic resin become popular since the ligno-cellulosic fibers are inexpensive, possess sufficient mechanical properties and environmental friendly due to their biodegradation properties. In tropical countries, plenty of abundance lignocellulosic fibers like jute, hemp, ramie, sisal, okra fibers, Palm Oil fiber etc. are grown and most of them have been employed for reinforcements in polymer matrices (Albano et al. 1999; Suardana et al. 2011; Khan et al. 2015b; Khan et al. 2016, Ramez A. Al-Mansob et al. 2013; Islam & Ani 2000). Among the lignocellulosic fibers, jute fiber is promizing on accunt of availability, low densities, nonabrasive nature, high specific properties, high modulus and renewable nature. Jute (Corchorus olitorius) is one of the most common agricultural fibers is cultivated almost exclusively in Bangladesh. Therefore, due to their avilability and suitable properties, jute fiber has the right potential for usage in composites.

Polypropylene (PP) is extensively used in engineering plastic materials for tremendous mechanical properties. It also has advantages like economy (price), ecological (recycling behaviors) and higher thermal stability and the effectiveness on filler reinforced composite. However, during impregnation of PP by lignocellulose fiber, interface shows incompatibility i.e. hydrophilic lignocellulose fiber cannot mix properly with hydrophobic PP. Therefore, afford need to reduce incompatibility on the interfacial adhesion between filler and matrix polymers for manufacturing high value composite materials.

A lot of conventional methods have practised to improve the interfacial adhesion of composites such as modifying the fber surface before composite fabrication, using compatibilizer during moulding or matix modification. The use of silane coupling agents, grafing by bifunctional monomers and the plasma treatment of the fiber surfaces are the most common techniques of interface modification of composites (Pothan et al. 2002; Khan et al. 2009; Khan et al. 2013a; El-Sabbagh 2014; Khan et al. 2015a; Khan et al. 2015b). In the present investigation, to improve the fiber matrix interaction, jute fiber surface have been modified by NaOH, dicumyl peroxide, potassium dicromate, and hydrogen peroxide. MAPP and DgPP have also used to improve interfacial properties which are well known compatibilizers for filler and matrix. It has been reported methane protons of Isotactic PP is often caused oxidative degradation. The degradation reaction proceed by a free radical chain reactive mechanism and formed carbonyl group (C = O) as well as hydroperoxide group (ROOH) (Alam et al. 2002). Another study used oxidative degraded PP (DgPP) as compatibilizer of cellulose/PP composite (Miyazaki et al. 2008).

## EXPERIMENTAL DESIGN

### MATERIALS

The water retted jute fiber (*Corchorus olitorius*) was collected from Kushtia district, Bangladesh. The polypropylene (PP) was obtained from Polyolefin Company (Singapore) Pvt. Ltd. in the form of pellets. Maleated polypropylene (MAPP) was purchased from Sigma-Aldrich in the granule form and degraded polypropylene (DgPP) was prepared in a crucible allowed by heating at 130°C for 20 h in presence of air. All analytical reagent (AR) grade chemicals were used in the investigation.

### FIBER PROCESSING

Fibers were cut into 25-30 cm and then scoured by (6.5 g/ℓ soap and 3.5 g/ℓ of $Na_2CO_3$) solution at 70-75°C during 30 min. The fiber to solution weight ratio was maintened at 1:50. After scouring the fiber was several times by distilled water and dried in the open air (Mondal and Khan 2008). The dried fibers were chemically treated with 4 different chemicals, namelyNaOH, dicumyl peroxide (DCP), $K_2Cr_2O_7$ and $H_2O_2$.

The dried jute fibers were immersed in 10 w/v% NaOH solution at 30°C maintained the fiber to liquor ratio at1:20 for 2 h. Then the fibers were washed repeatedly by distilled water and finally washed by dilute acetic acid to remove NaOH sticking to the fiber surface. The neutralize fibers were dried in air and finally stored in desiccator (Khan et al. 2015b).

6 w/v% dicumyl peroxide (DCP) solution was prepared with acetone. The NaOH treated fiber was socked in the DCP solution for 30 min at 30°C maintained the fiber to liquor ratio at 1:11. The treated fiber was washed by distilled water and dried at room temperature. The NaOH treated fiber were treated by 0.055 w/v% $K_2Cr_2O_7$ solution maintained the fiber to liquor ratio 1: 15 for 30 min at 60°C with occasionally added few drops of $H_2SO_4$. Then the fiber was washed by distilled water and dried in air (Khan et al. 2015b).

The jute fibers were treated by 5 w/v% $H_2O_2$ solution maintained the fiber to liquor ratio 1: 80 for 2 h at 90°C. During the treatment the pH of adjusted by adding 7-8 drops of 0.2 M $CH_3COOH$ solution. The fiber was filtered and washed by distilled water. Then the fibers were immersed in 0.2% $Na_2S_2O_5$ solution maintained the fiber to liquor ratio 1:20 for 15 min at 30°C. Finally the fiber was washed and dried in air and finally stored in desiccator (Li & Wang 2013).

TABLE 1. Various types of composite sample prepared in different parameters

| Sample | Fiber chemical treatments | Fiber wt% | Matrix | Coupling agent | Composite |
|---|---|---|---|---|---|
| A | No chemical treatment | 30% | PP (70 wt%) | - | Raw JF + PP |
| B | No chemical treatment | 30% | PP | DgPP (2 wt%) | Raw JF + PP + DgPP |
| C | No chemical treatment | 30% | PP | MAPP (2 wt%) | Raw JF + PP + MAPP |
| D | 10 w/v% NaOH | 30% | PP (70 wt%) | - | 10% NaOH JF + PP |
| E | 10 w/v% NaOH | 30% | PP (68 wt%) | DgPP (2 wt%) | 10% NaOH JF + PP + DgPP |
| F | 10 w/v% NaOH | 30% | PP (68 wt%) | MAPP (2 wt%) | 10% NaOH JF + PP + MAPP |
| G | 6 w/v% DCP | 30% | PP (70 wt%) | - | 6% DCP JF + PP |
| H | 6 w/v% DCP | 30% | PP (68 wt%) | DgPP (2 wt%) | 6% DCP JF + PP + DgPP |
| I | 6 w/v% DCP | 30% | PP (68 wt%) | MAPP (2 wt%) | 6% DCP JF + PP + MAPP |
| J | 0.055 w/v% $K_2Cr_2O_7$ | 30% | PP (70 wt%) | - | 0.055% $K_2Cr_2O_7$ JF + PP |
| K | 0.055 w/v% $K_2Cr_2O_7$ | 30% | PP (68 wt%) | DgPP (2 wt%) | 0.055% $K_2Cr_2O_7$ JF + PP + DgPP |
| L | 0.055 w/v% $K_2Cr_2O_7$ | 30% | PP (68 wt%) | MAPP (2 wt%) | 0.055% $K_2Cr_2O_7$ JF + PP + MAPP |
| M | 5 w/v% $H_2O_2$ | 30% | PP (70 wt%) | - | 5% $H_2O_2$ JF + PP |
| N | 5 w/v% $H_2O_2$ | 30% | PP (68 wt%) | DgPP (2 wt%) | 5% $H_2O_2$ JF + PP + DgPP |
| O | 5 w/v% $H_2O_2$ | 30% | PP (68 wt%) | MAPP (2 wt%) | 5% $H_2O_2$ JF + PP + MAPP |

### COMPOSITE PREPARATION

The fiber bundles were cut into approx. 10 cm length, and then placed in an air oven for an week to remove the fiber surface moisture. Virgin PP sheet was prepared in an open S-S mold. Mold was electrically heated by thermostat at 180°C for 115 min. Then the mold was placed in hydraulic press and pressure was set to 50 kN. the mold was then cooled by cold tap water flow and transparent PP sheet was obtained. The PP sheet were cut into 10 × 10 $cm^2$ size and placed in a closed mold with same dimension. The predetermined amount fibers were unidirectionaly sprayed on the PP sheet (Table 1). The fixed amount of MAPP or DgPP was added as compatibilizer. The fibers were then covered by PP sheets. The mold was put in between two hot plates of compression molding machine. Pressure and temperature were fixed to 50 kN and 180°C respectively. The heating was gradually increased upto 180°C. After 30 min, heating was stopped and mold was cooled by tap water. Finally, the composite sample was taken out from mold and cut for the mechanical testing.

## MEASUREMENTS

Tensile properties were measured using Universal Testing Machine (Hounsfield UTM 10 kN) by ASTM standard D638 methods. The crosshead speed was 50 mm/min and gage length was 50 mm. The results are taken from the average of 10 measurements. The water absorption property was measured by ASTM D570 method . The specimens were dried in an oven at 80°C for 24 h prior to testing. Then those samples were soaked into water at room temperature.

The composites were taken out from water after every 24 h and all surface moisture was removed with tissue paper. The weight gain was calculated using Equation 1:

$$\text{Water absorption } \% = \frac{(W_t - W_o) \times 100}{W_o} \quad (1)$$

where $W_o$ and $W_t$ are the initial weight and weight after water absorption, respectively. The data reported are average value obtained from ten separate samples of each composite. The thermal properties of composites were assessed by thermo-gravimetric analyzer supplied by TA Instrument (EXTAR 6000 STATION, Seiko Instrument, Inc. Japan). About 20 mg of composite sample was taken for each analysis. The heating was increased by the rate of 20°C/min from 25°C to 600°C in nitrogen environment (gas flow rate 50 ml/min).

## RESULTS AND DISCUSSION

The surface of jute fiber has been chemically modified through different chemical process. The performance of DgPP and MAPP coupling agents at interface modification of composites were investigated in terms of their water absorption, mechanical and thermal properties.

### WATER ABSORPTION PROPERTIES

Figure 1 shows the effect of immersion time on water absorption of raw and treated jute fiber PP composites. Raw JF + PP composite (Sample A) are taken as control for comparison. It shows that the lowest water absorption is found in case of 10% NaOH JF + PP + MAPP composite and water absorption is increased with the increase of immersion time. It is also observed that, the addition of coupling agents MAPP and DgPP in composites give lower water absorption than JF + PP composite. With the addition of MAPP and DgPP coupling agents in composites, the percentage of water absorption is decreased. It may be due to the enhancement of surface adhesion of fibers and PP matrix that reduced the water consumption in the interfacial voids. Probably the coupling agents took part on the esterification reaction with the -OH group in jute fiber (Khan et al. 2013a).

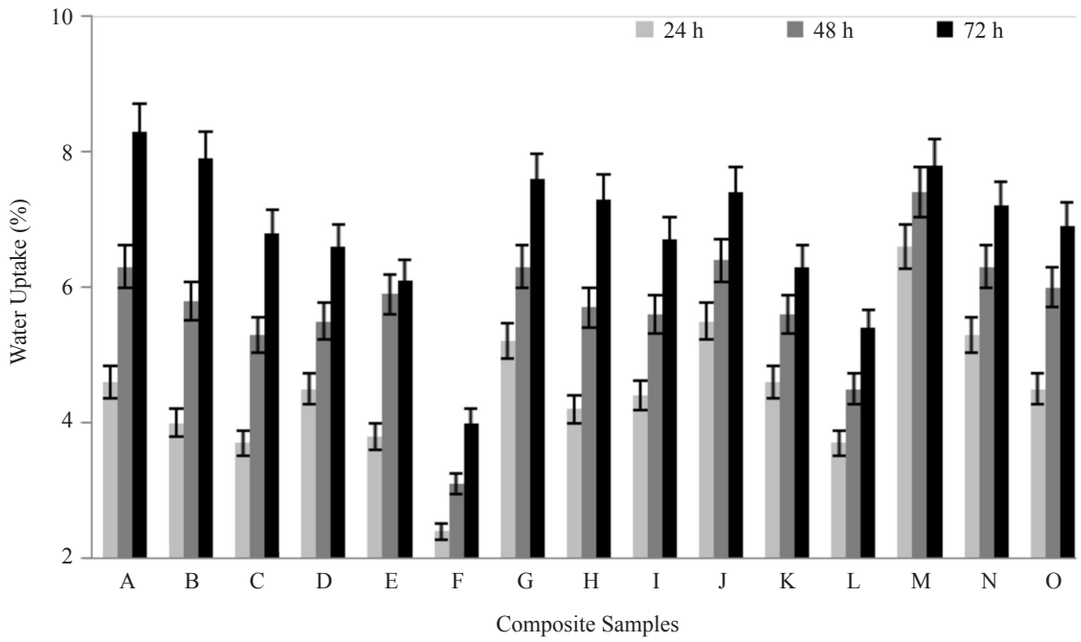


FIGURE 1. Effect of immersion time on water absorption of raw and treated jute fibers PP composites

### MECHANICAL PROPERTIES

The tensile properties of unidirectional jute fiber reinforced PP composites were investigated by means of the effect of coupling agents (DgPP, MAPP) and chemical treatments of fiber. The tensile strength, Young's modulus and elongations were obtained from tensile stress-strain data.

Figure 2 shows the effect of chemical treatments and coupling agents (DgPP and MAPP) on tensile strength (TS) of raw and treated jute fibers PP composites. It is observed that the coupling agents added in both raw and treated jute fibers – PP composites yield higher TS than without coupling agents. It has also been revealed that all chemically treated fiber composites give higher TS raw fiber composite. Among

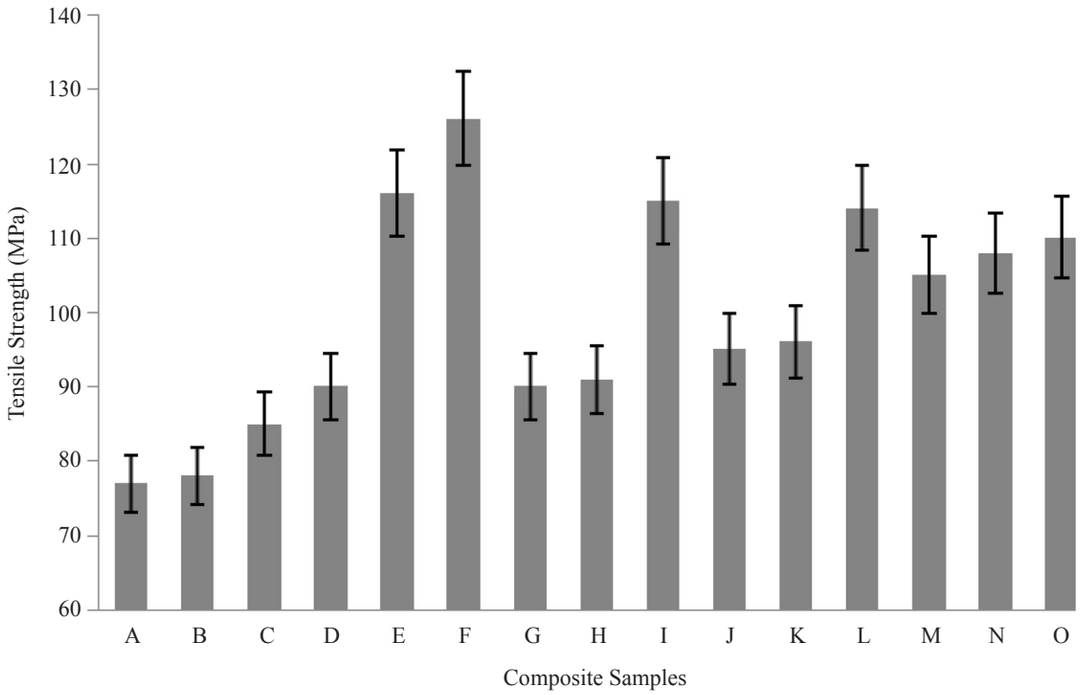


FIGURE 2. The effect of chemical treatments and coupling agents on the tensile strength of raw and treated jute fiber PP composites

the composites, highest value of TS is found for 10% NaOH JF + PP + MAPP composite and the lowest value is obtained for raw JF + PP composite. The chemical treatments are employed in this investigation such as NaOH, DCPO, $K_2Cr_2O_7$, $H_2O_2$ are all oxidative treatments process. These treatments can make the fiber surface become 'clean' due to removal of impurities, pectin, waxes, hemicellulose and part of lignin. The removal of these substances enhances the surface roughness. Therefore, the jute fiber easily wetted by PP matrix. In addition, interlocking between fiber and matrix is increased. The result shows, the presence of coupling agents caused better adhesion of jute fibers and polypropylene matrix and forms strong interfacial bond between them. The coupling agents MAPP and DgPP have similar molecular arragment with extra functional groups which forms chemical bond with hydrophilic jute fiber. The main PP chain of the coupling agents entangle the mutual chains of PP and therefore builds stronger interfacial adhesion between the PP matrix and jute fiber (Khan et al. 2013a; Miyazaki et al. 2008; Khan et al. 2015a). Both coupling agents have similar mechanism of bond formation via esterification between the -OH group in jute fiber and the reactive γ-lactone groups in the coupling agents (Khan et al. 2013a).

Figure 3 shows the comparison of the effect of chemical treatments and coupling agents on YM of raw and treated jute fibers PP composites. It shows that the addition of coupling agents increases YM of raw and treated jute fibers PP composites. The highest and lowest values of YM are obtained for 10% NaOH JF + PP + MAPP and raw JF + PP composites, respectively.

Figure 4 shows the effect of chemical treatments and coupling agents on elongation properties (PE) of raw and treated jute fibers PP composites. From the Figure 4, it is observed that the highest PE value was found for raw JF + PP composite, then 10%NaOH JF + PP + MAPP and the lowest for 0.05% $K_2Cr_2O_7$ JF + PP + MAPP composites. The lower values of elongation for composites which were made by using chemical treated fiber and coupling agents may be due to the increase of brittleness of the composite.

## THERMAL PROPERTIES

Figure 5(A) shows the Thermogravimetry (TG), Differential Thermogravimetry (DTG) and Differential Thermal Analysis (DTA) curves of raw JF + PP, raw JF + PP + DgPP, raw JF + PP + MAPP composites. TG curve shows two steps degradation, first is related to fiber degradation and the second is related to polymer polypropylene (PP) degradation. The Figure 5(A) shows that raw JF + PP composite has slightly lower onset and decomposition temperature than raw JF + PP + DgPP composite but slightly higher than raw JF + PP + DgPP composite. The initial loss due to moisture are 2.4% for raw JF + PP, 0.8% for raw JF + PP + DgPP, 1.9% for raw JF + PP + MAPP composite.

All DTA curves show three different endothermic peaks. First, second, third peaks are related to melting of PP, fiber degradation and PP degradation respectively. DTG curves of raw JF + PP, raw JF + PP + DgPP, raw JF + PP + MAPP composites show two peaks each. First and second peaks are related to fiber and PP polymer degradation respectively.

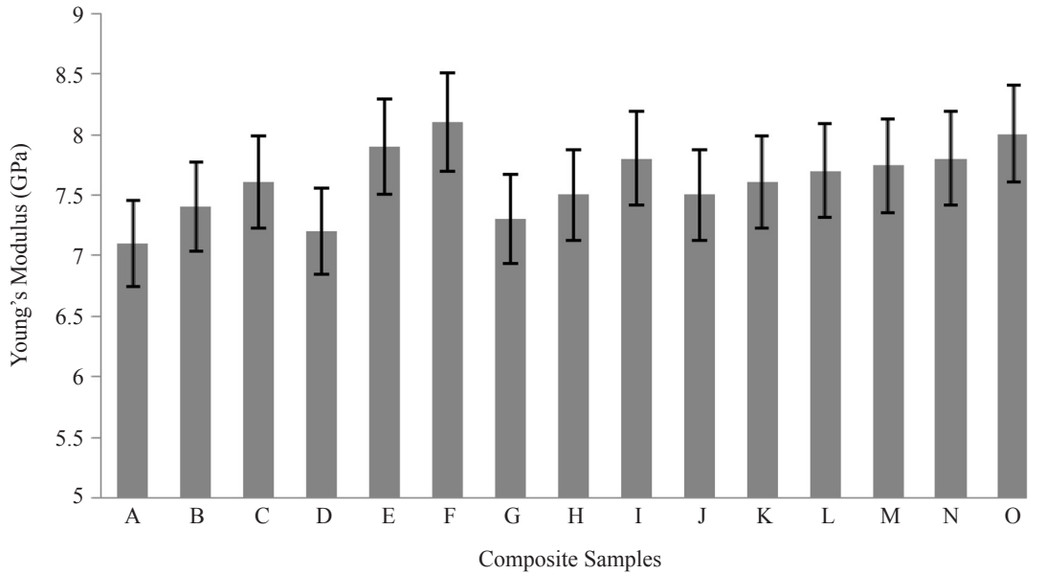


FIGURE 3. The effect of chemical treatments and coupling agents on Young's modulus of raw and treated jute fiber PP composites

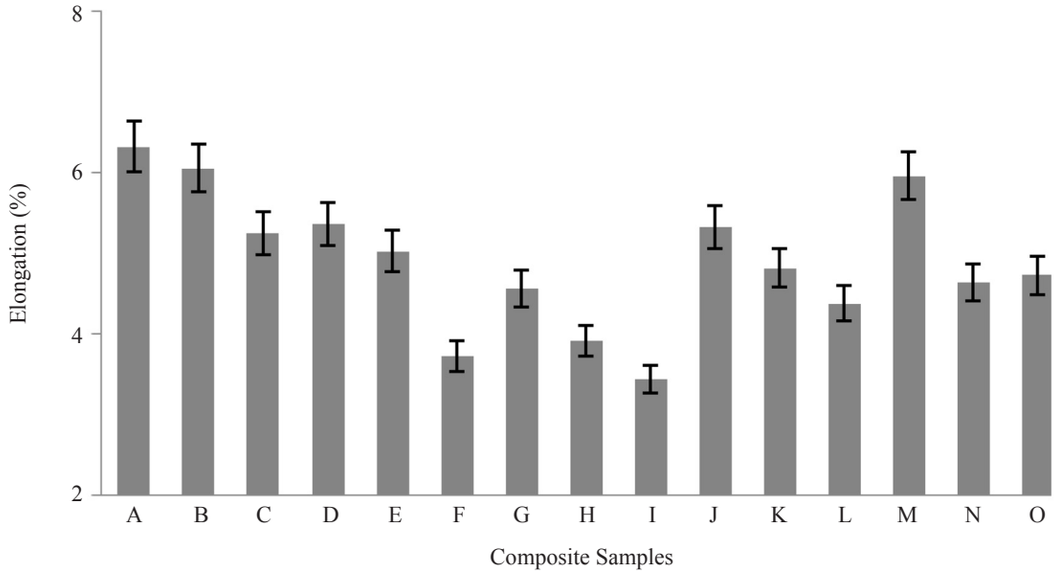


FIGURE 4. The effect of chemical treatments and coupling agents on elongation at break of raw and treated jute fiber PP composites

Figure 5(b) and 5(c) shows the TG, DTA & DTG curves for 5% $H_2O_2$, 0.05% $K_2Cr_2O_7$, 6% DCPO & 10% NaOH JF + PP + DgPP and 5% $H_2O_2$, 0.05% $K_2Cr_2O_7$, 6% DCPO & 10% NaOH JF + PP + MAPP composites, respectively. No detectable change is observed for all types of composites.

Table 2 shows the properties of sawdust reinforced polypropylene composities of some previous research. Since the properties of composites mostly depends on the properties of matrix use on it. The improvement of mechanical properties of our composites is much more higher than others. On the other hand, a great extent of water uptake also found in this study.

## CONCLUSION

The study illustrate the properties of composites fabricated by hot press molding method using polypropylene (PP) as matrix and jute fibers. The 2 wt% DgPP and MAPP are used as compatibilizer to improve the surface adhesion between the hydrophilic jute fibers and hydrophobic PP matrix. As a result, composites showed higher mechanical properties than that of composites prepared by without coupling agent. In case of, the raw JF + PP composite have higher ability to absorb water and 10%NaOH JF + PP + MAPP composite have lower ability to absorb water. No remarkable changes found on thermal degradation of raw and treated fibers reinforced composites which mean that fibers are not degraded during chemical treatments.

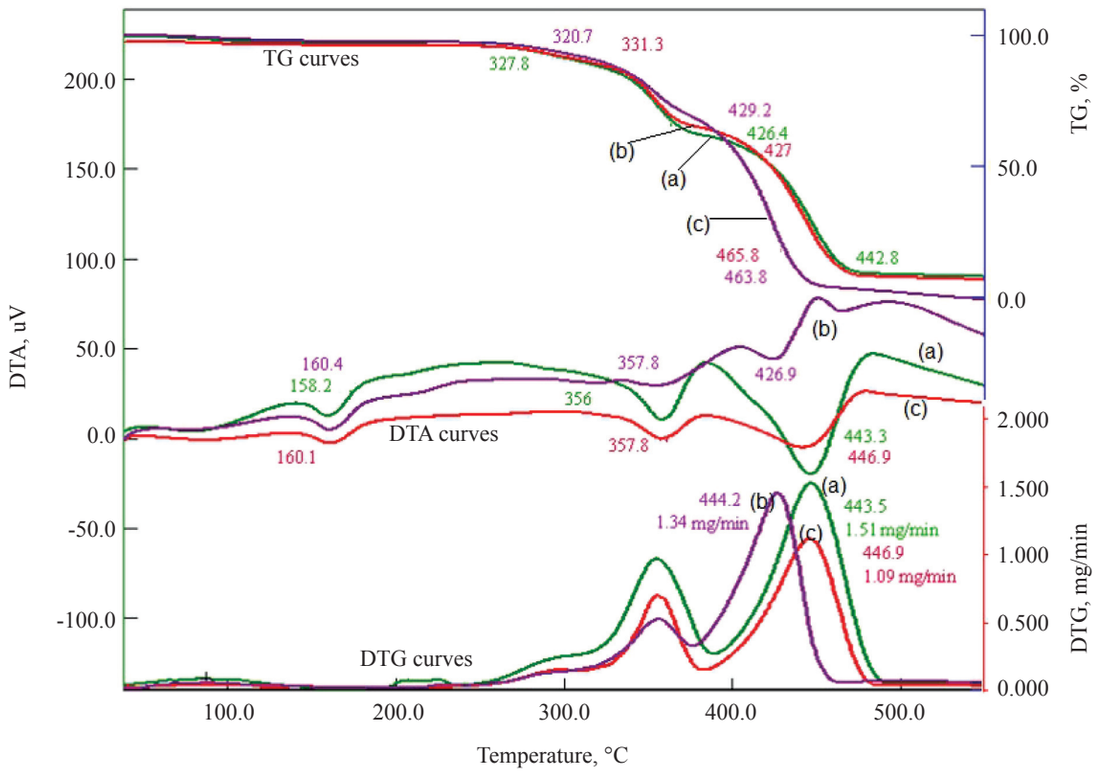


FIGURE 5(a). DTA, TG & DTG curves of (a) raw JF + PP, (b) raw JF + PP + DgPP and (c) raw JF + PP + MAPP composites

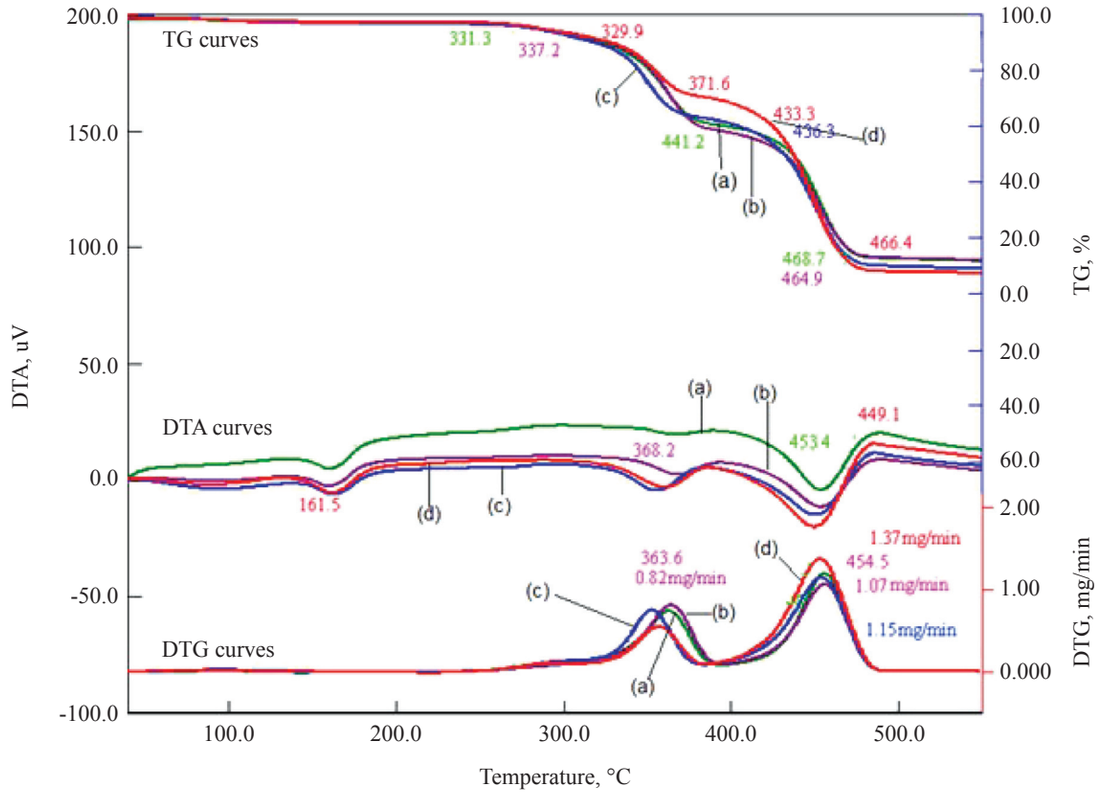


FIGURE 5(b). TG, DTA & DTG curves of (a) 5% $H_2O_2$, (b) 0.05% $K_2Cr_2O_7$, (c) 6% DCPO and (d) 10% NaOH JF + PP + DgPP composites

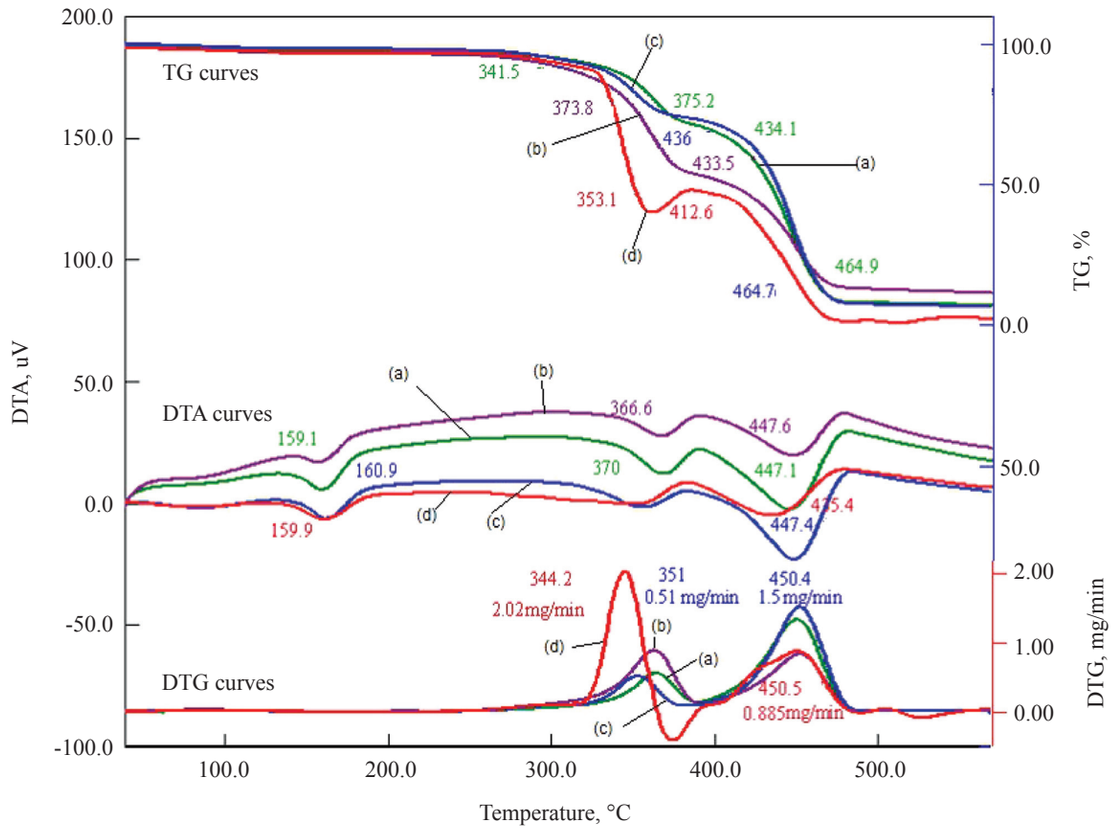


FIGURE 5(c). TG, DTA & DTG curves of (a) 5 w/v% $H_2O_2$, (b) 0.05 w/v% $K_2Cr_2O_7$, (c) 6 w/v% DCPO and (d) 10 w/v% NaOH JF + PP + MAPP composites

TABLE 2. Comparative result from some previous study

| Amount of Sawdust | Chemical treatments | Compatibilizer | Method of Molding | Tensile strength (MPa) | Tensile Modulus (GPa) | Water absorption (%) | Ref. |
|---|---|---|---|---|---|---|---|
| 15 wt% | Diazonium salt | | Injection | 28.4 | 1.2 | 0.2 | Rahman et al. 2010 |
| 40 wt% | | DCP or BPO peroxide/maleic anhydride (MAH) | Compression | 24.8 | - | - | Santos et al. 2016 |
| 50 wt% | 3-aminopropyltriethoxysilane | MAH | Injection | 49 | - | - | Kim et al. 2010 |
| | 3-methacryloxypropyltrimethoxysilane (MPS) | MAH | Injection | 49 | - | - | |
| | vinyltrimethoxysilane | MAH | Injection | 49.2 | - | - | |
| 30 wt% | NaOH | MAPP | compression | 127 | 8.1 | 2.1 | |
| | NaOH | DgPP | compression | 118 | 7.9 | 3.9 | |

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REFERENCES

Alam, M.S., Nakatani, H., Ichiki, T., Goss, G.S., Liu, B. & Terano, M. 2002. Improvement of the photostability of isotactic polypropylene by the incorporation of ethylene. *J Appl Polym Sci* 86: 1863-1867.

Albano, C., González, J. Ichazo, M. & Kaiser, D. 1999. Thermal stability of blends of polyolefins and sisal fiber. *Polym Degrad Stab* 66: 179-190.

Al-Mansob, R.A., Ismail, A., Algorafi, M.A., Hafezi, M.H. & Baghini, M.S. 2013. Comparison between mixtures of asphalt with palm oil shells and coconut shells as additives. *Jurnal Kejuruteraan* 25: 25-31.

Dos Santos, L.P., Trombetta, E., Flores-Sahagun, T.S. & Satyanarayana, K.G. 2016. Effect of domestic compatabilizer on the performance of polypropylene-sawdust composites. *J Compos Mater* 50: 1353-1365.

El-Sabbagh, A. 2014. Effect of coupling agent on natural fibre in natural fibre/polypropylene composites on mechanical and thermal behaviour. *Compos Part B Eng* 57: 126-135.

Islam, M. & Ani, F. 2000. Production of bio-oil from Palm Oil Shell. *Jurnal Kejuruteraan* 12: 105-116.

Khan, G. M. A., Palash, S.R.S., Terano, M. & Alam, M.S. 2015a. Hybrid natural fibers/isotactic polypropylene composites with degraded polypropylene as compatibilizer. *Materials Chemistry and Physics* 1(2): 110-115.

Khan, G.M.A., Shaikh, H., Alam, M.S. & Gafur, M.A. 2015b. Effect of Chemical treatments on the physical properties of non-woven jute/PLA biocomposites. *BioResources* 10: 7386-7404.

Khan, G.M.A., Alam Shams, M.S., Kabir, M.R., Gafur, M. A., Terano, M. & Alam, M.S. 2013. Influence of chemical treatment on the properties of banana stem fiber and banana stem fiber/coir hybrid fiber reinforced maleic anhydride grafted polypropylene/low-density polyethylene composites. *J Appl Polym Sci* 128: 1020-1029.

Khan, G.M.A., Shaheruzzaman, M., Rahman, M.H., Abdur Razzaque, S.M., Islam, M.S. & Alam, M.S. 2009. Surface modification of okra bast fiber and its physico-chemical characteristics. *Fibers Polym* 10: 65-70.

Khan, G.M.A, Yilmaz, N.D. & Yilmaz, K. 2016. Okra bast fiber as potential reinforcement element of biocomposites: Can it be the flax of the future? In *Handbook of Composite from Renewable Materials*, edited by Thakur, V. K. Wiley Scrivener.

Kim, T.W., Lee, S.Y., Chun, S.J., Doh, G.H. & Paik, K.H. 2010. Effect of silane coupling on the fundamental properties of wood flour reinforced polypropylene composites. *J Compos Mater* 45: 1595-1605.

Li, X. & Wang, X.R. 2013. Effects of hydrogen peroxide treatment on structure and properties of natural color silk. *Adv Mater Res* 821-822: 90-93.

Miyazaki, K., Okazaki, N., Terano, M. & Nakatani, H. 2008. Preparation and characterization of cellulose/polypropylene composite using an oxidatively degraded polypropylene. *J Polym Environ* 16: 267.

Mondal, I.H.M. & Khan, G.M.A. 2008. Effect of acrylic monomers grafting onto jute constituents with potassium persulphate initiator catalysed by Fe (II). *Cellul Chem Technol* 42: 9-16.

Pothan, L., George, J. & Thomas, S. 2002. Effect of fiber surface treatments on the fiber–matrix interaction in banana fiber reinforced polyester composites. *Compos Interfaces* 9: 335-353.

Rahman, M.R., Islam, M.N. & Huque, M.M. 2010. Influence of fiber treatment on the mechanical and morphological properties of sawdust reinforced polypropylene composites. *J Polym Environ* 18: 443-450.

Suardana, N.P.G., Piao, Y. & Lim, J.K. 2011. Mechanical properties of hemp fibers and hemp/PP composites: Effects of chemical surface treatment. *Materials Physics and Mechanics* 11: 1-8.

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